



Docket No.: JCLA10633

PATENT

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant : P. Y. Young et al.  
Application No. :  
Filed : December 12, 2003  
Title : EMULSION TYPE MODIFIER FOR IMPROVING  
PROPERTIES OF PRESSURE SENSITIVE  
EMULSION ACRYLIC ADHESIVES

Grp./Div. :  
Examiner :

Docket No. : JCLA10633

**STATEMENT BY TRANSLATOR UNDER 37 C.F.R. § 1.52(d)**

Assistant Commissioner for Patents Post Office Box 7068  
Alexandria, VA 22313-1450

Commissioner:

I hereby, confirm that the English translation enclosed herewith is an accurate translation of above-captioned US Nonprovisional Application.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date

Feb. 16, 2004

By

Caroline Pao



FILE: 10633USF.RTF

EMULSION TYPE MODIFIER FOR IMPROVING PROPERTIES OF PRESSURE  
SENSITIVE EMULSION ACRYLIC ADHESIVES

5 CROSS-REFERENCE TO RELATED APPLICATION

This application claims the priority benefit of Taiwan application serial no.  
91135910, filed on December 12, 2002.

BACKGROUND OF THE INVENTION

10 Field of Invention

[0001] The present invention relates to an emulsion type modifier. More particularly,  
the present invention relates to an emulsion type modifier for improving properties of  
the pressure-sensitive emulsion acrylic adhesives.

Description of Related Art

15 [0002] In general, the pressure-sensitive emulsion acrylic adhesives have inferior  
surface adhesion toward the low surface energy substrates, such as the polyolefins. Thus,  
tackifiers are usually needed for improving the adhesion of the pressure-sensitive  
emulsion acrylic adhesives. The representative tackifiers for the pressure-sensitive  
emulsion acrylic adhesives are the rosin resins. The rosin resins are in fact one type of  
20 acidic natural resins extracted from the pine trees. However, the quality of the rosin  
resins is not always stable and usually varies if obtained from different pine trees of  
various origins, from diverse weathers or numerous tree species. Due to the compatibility  
of the rosin resins and the pressure-sensitive emulsion acrylic adhesives, the physical

properties of the pressure-sensitive emulsion acrylic adhesives may be deteriorated if too much rosin resins are added.

#### SUMMARY OF THE INVENTION

5 [0003] The present invention provides an emulsion type modifier of high stability and good durability, of which a small quantity of addition significantly improves adhesion of the pressure-sensitive emulsion acrylic adhesives.

[0004] As embodied and broadly described herein, the present invention provides an emulsion type modifier for improving properties of the pressure-sensitive emulsion acrylic adhesives. The emulsion type modifier comprises: (a) 15 to 35 parts per hundred of monomers of a C6-C12 diester of a dicarboxylic acid; (b) 15 to 35 parts per hundred of monomers of C2-C16 vinyl esters; (c) 35 to 65 parts per hundred of monomers of C4-C8 alkyl acrylates; based on the total weight of (a)+(b)+(c), and (d) 0.1 to 2.0 parts per hundred of an acetoacetoxy functional monomer; (e) 0.1 to 2.0 parts per hundred of a nonionic surfactant; and (f) 0.4 to 3.0 parts per hundred of an anionic surfactant. The emulsion type modifier of the present invention has an average particle diameter between about 0.1~0.4 microns, a glass transit temperature (Tg) between about -50~-30°C and a solid content of about 50%-70% by weight (wt.%).

[0005] By adding the emulsion type modifier of the present invention, the steel adhesion, polyethylene (PE) adhesion, the loop tack and the holding power of the pressure-sensitive emulsion acrylic adhesives can be improved. The addition amount of the emulsion type modifier is about 5 to 30 parts per hundred, relative to the amount of the pressure-sensitive emulsion acrylic adhesive.

[0006] It is to be understood that both the foregoing general description and the following detailed description are exemplary, and are intended to provide further explanation of the invention as claimed.

5 DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0007] The present invention provides an emulsion type modifier for improving the steel adhesion, polyethylene (PE) adhesion and the loop tack and holding power of the pressure-sensitive emulsion acrylic adhesives.

[0008] The emulsion type modifier can be produced by any available method of  
10 emulsion and polymerization. According to the preferred embodiment, the composition of the emulsion type modifier comprises monomers of:

(a) a diester of a dicarboxylic acid containing 4-12 carbons, for example, dioctyl maleate or dibutyl maleate, 15 to 35 parts per hundred;

(b) vinyl ester containing 2-16 carbons, for example, vinyl butyrate, allyl acetate,  
15 vinyl isobutyrate or vinyl acetate, 15 to 35 parts per hundred;

(c) alkyl acrylates containing 4-8 carbons, for example, ethyl acrylate, butyl acrylate, 2-ethyl hexyl methacrylate, decyl acrylate, methyl methacrylate or butyl methacrylate, 35 to 65 parts per hundred; and

(d) monomers containing acetoacetoxy functional groups, for example, ethyl  
20 acetoacetoxy methacrylate or butyl acetoacetoxy methacrylate, 0.1 to 2.0 parts per hundred, based on the total weight of (a)+(b)+(c).

[0009] The composition of the emulsion type modifier may further comprise different types of surfactants for improving stability and reaction rates for emulsion and polymerization, and film build and hydrolysis resistance of the products. The surfactants

include a mixture of anionic surfactants and nonionic surfactants, 0.5 to 5.0 parts per hundred, based on the total weight.

[0010] The composition of the emulsion type modifier may further comprise:

(e) a nonionic surfactant, for example, polyoxethylene nonyl phenyl ether, polyoxethylene octyl phenyl ether, polyethylene glycol alkyl phenol ether, diallyl aliphatic acid ester, or polyethylene glycol diallyl aliphatic acid ester, 0.1 to 2.0 parts per hundred based on the total weight;

(f) an anionic surfactant, for example, sodium dodecylbenzosulfonate, sodium dodecylsulfonate, disodium dodecyl sulfonate phenyl ether, disodium n-stearyl succinate sulfonate or ammonium polyoxethylene nonyl phenyl ether sulfate, 0.4 to 3.0 parts per hundred based on the total weight; and

(g) de-ionized water, 60 to 70 parts per hundred.

[0011] The monomer composition of the emulsion type modifier begin to react with each other through at least one initiator with the addition amount of 0.1 to 2.0 parts per hundred based on the total weight of (a)+(b)+(c). For example, the initiator can be sodium persulfate, ammonium persulfate or potassium persulfate. With the existence of the surfactant and the initiator, the monomers in the reaction tank begin to react under heating to form latex particles, under reaction temperature 50°C-90°C and the reaction time of 5-10 hours. The monomers of C4-C12 diester of a dicarboxylic acid, the monomers of C2-C16 vinyl esters, the monomers of C4-C8 alkyl acrylates, the acetoacetoxy monomers, the nonionic surfactant, the anionic surfactant and the initiator can be added simultaneously or sequentially in batches. The emulsion type modifier of the present invention has an average particle diameter between about 0.1~0.4 microns,

a glass transit temperature (T<sub>g</sub>) between about -50~-30°C and a solid content of about 50%-70% by weight (wt.%).

[0012] The present invention provides a pressure-sensitive adhesive and the composition of the pressure-sensitive adhesive includes 100 parts per hundred of pressure-sensitive emulsion acrylic adhesive and 5-30 parts per hundred of the  
5      aforementioned emulsion type modifier. The mixing ratio of the pressure-sensitive emulsion acrylic adhesive and the emulsion type modifier can be adjusted based on the requirements. Relative to the amount of the pressure-sensitive emulsion acrylic adhesive, the emulsion type modifier is added in 5-30 parts per hundred, for effectively  
10     improving the steel adhesion, PE adhesion, the loop tack and the holding power of the pressure-sensitive emulsion acrylic adhesives (referring to Table 3).

#### Preparation of the emulsion type modifier

##### Example 1

[0013] 44g of de-ionized water and 0.25g NH<sub>4</sub>.polyoxethylene nonyl phenyl ether  
15     sulfate (Chun-Zih Synthesis Inc.) are consistently mixed to obtain the solution A.

[0014] 76.7g of de-ionized water, 2.9g of polyoxethylene nonyl phenyl ether (Chun-Zih Synthesis Inc.), 12.9g of NH<sub>4</sub>.polyoxethylene nonyl phenyl ether sulfate, 157.3g of 2-ethyl hexyl methacrylate monomers, 102.6g of dioctyl maleate monomers, 85.5g of vinyl acetate monomers and 1.0g of ethyl acetoacetoxy methacrylate monomers are  
20     consistently mixed to obtain the solution B.

[0015] 87.5g of de-ionized water, 1.8g of sodium bicarbonate and 0.9g of potassium persulfate are mixed to prepare the solution C.

[0016] 20.5g of de-ionized water and 0.9g of potassium persulfate are mixed to prepare the solution D. Preparation of the solutions A, B, C and D can be performed individually.

[0017] The solution A and the half quantity (1/2) of the solution D are poured in to the emulsion and polymerization reaction system, including the stirrer, the thermometer, the heater, the reflux condenser and the five neck flask. When the solution is heated to 70°C, the mixture of 1/10 the solution B and 1/2 the solution D is steadily fed within 20-30 minutes into the emulsion and polymerization reaction system. Then heated to 80°C, the remaining solution B and the solution C are slowly fed within 240 minutes into the emulsion and polymerization reaction system. Next, the reaction temperature is increased to 85°C and reacted for 90 minutes, and then cooled to the room temperature. The resultant emulsion type modifier has a solid content of 59.5%, pH 5.2, viscosity 23000cps and its physical properties are shown in Table 1.

#### Example 2

[0018] The preparation processes are performed as Example 1, except replacing 2.9g of polyoxethylene nonyl phenyl ether in the solution B with 2.6g of polyoxethylene octyl phenyl ether (Chun-Zih Synthesis Inc.). The resultant emulsion type modifier has a solid content of 59.0%, pH 5.1, viscosity 22800cps and its physical properties are shown in Table 1.

[0019] Table 1 Physical properties of the products

Physical properties	Example 1	Example 2
Particle size ( $\mu$ m)	0.16	0.15
Solid content (%)	59.5	59.0
Viscoity (cps)	23000	22800
Steel adhesion (kg/in)	1.70	1.65
PE adhesion (kg/in)	1.20	1.15
Loop tack (kg/in)	1.70	1.68
Holding power (min)	8000	7800

[0020] Gluing conditions: the thickness of the polyester film is 25  $\mu$  m; the thickness of the glue is 22  $\mu$  m.

- 5 [0021] Table 2 Physical properties and adhesion for pressure-sensitive emulsion acrylic adhesive (Shi-Mei Inc.; PS-8105)

Physical properties	Pressure-sensitive emulsion acrylic adhesive (Shi-Mei Inc.; PS-8105)
Solid content (%)	60.0
Viscoity (cps)	100
Steel adhesion (kg/in)	0.96
PE adhesion (kg/in)	0.26
Loop tack (kg/in)	1.30
Holding power (min)	630

[0022] Coating conditions: the thickness of the polyester film is  $25\ \mu\text{m}$ ; the thickness of the adhesive is  $22\ \mu\text{m}$ .

[0023] The physical properties listed in Tables 1 and 2 are obtained through the following processes:

5           1. Solid content (%): after measuring the weight of the plate (a), 0.5g of latex (b) is placed on the plate and both are put into the oven, under  $150^{\circ}\text{C}$  for 1 hour. Then the plate and latex are weighted (c). The calculation formulation:  $\text{solid content (\%)} = \{(c - a)/b\} * 100$ .

10           2. Viscosity (cps): measured by Brook field viscosimeter,  $25^{\circ}\text{C}$ , 4# rotating rod/30rpm.

          3. Steel adhesion (kg/in): according to ASTM D-1000 standards.

          4. PE adhesion (kg/in) : according to ASTM D-1000 standards.

15           5. Loop tack (kg/in): the sample size of  $25 \times 200\text{mm}$ , the glued surface facing downwards and both ends being overlapped 25mm to form a loop. The glued surface is in contact with the steel plate and the tape is moving up in a speed of  $300\text{mm/min}$ , so as to calculate the maximum adhesion when the tape is separate from the steel plate, units in kg/in.

20           6. Holding power (min): according to PSTC-7 tape testing standards, the sample size of  $25 \times 25\text{mm}$ , glued to the steel plate and holding for 1kg. The retaining time is recorded, units in minutes.

The effect tests:

[0024] The product of example 1 is mixed with the pressure-sensitive emulsion acrylic adhesive (Shi-Mei Inc.; PS-8105) in different ratios and then spin coated to the polyester film (the polyester film thickness  $25\ \mu\text{m}$ ; the adhesive thickness  $22\ \mu\text{m}$ ).

After dried under 80°C/5 minutes, it is placed under 25°C/65RH (relative humidity) for 24 hours and then the steel adhesion, PE adhesion, the loop tack and the holding power are measured. The results are listed in Table 3.

[0025] Table 3 Effect tests

Physical properties	Example 1 / pressure-sensitive emulsion acrylic adhesive (phr)			
	0/100	5/100	20/100	30/100
Steel adhesion (kg/in)	0.96	1.16	1.20	1.35
PE adhesion (kg/in)	0.26	0.45	0.78	1.12
Loop tack (kg/in)	1.30	1.36	1.49	1.50
Holding power (min)	630	2500	3410	5890

5

[0026] Coating conditions: the thickness of the polyester film is 25  $\mu$  m; the thickness of the adhesive is 22  $\mu$  m.

[0027] From Table 3, it is shown that the adhesion, including steel adhesion, polyethylene (PE) adhesion, the loop tack and the holding power, of the pressure-sensitive emulsion acrylic adhesives can be effectively improved by adding the emulsion type modifier of the present invention.

[0028] It will be apparent to those skilled in the art that various modifications and variations can be made to the structure of the present invention without departing from the scope or spirit of the invention. In view of the foregoing, it is intended that the present invention cover modifications and variations of this invention provided they fall within the scope of the following claims and their equivalents.